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## -- NOTES --

# POPULATION INVERSION OF A. U. V. ATOMIC LINE IN RECOMBINING PLASMA NOZZLE FLOW*

S.W. Bowen** and C. Park†

## Abstract

The self-absorption of the neutral carbonline at 2478.6 Å ($2p^2\ ^1S_0 - 2p\ 3s\ ^1P_1{}^o$) has been experimentally measured by placing a concave mirror behind expanding, high pressure, arc heated plasmas issuing from a 1.27 cm constricted arc tunnel. Negative absorption coefficients indicating population inversions have been observed in both helium-methane and argon-methane plasmas. To date the largest absorption coefficient has been $\kappa_0{}^* \approx -.292\ \text{cm}^{-1}$ using a mole fraction of carbon $= .033$ in a helium-methane mixture, with total mass flow 2.5 gm/sec, 4.75 atm cathode pressure and enthalpy ≈ 190 Mj/Kg. The effective area ratio at the observation station was ≈ 50.

## Introduction

Recent theoretical calculations[1] and experimental measurements[2] have indicated that a large overpopulation of the upper neutral atom excited states with respect to the ground state can exist in an arc-heated plasma expanding through a nozzle. In Ref. 2 it is shown experimentally that such an overpopulation results ultimately in a population inversion between a pair of states corresponding to a visible or infra-red spectral line. In Ref. 1 it is predicted that a population inversion may occur also for an ultraviolet line. The purpose of the present work is to examine experimentally the population inversion of an ultraviolet line. Here, spectroscopic measurements made on high pressure, high temperature arc-heated helium-methane or argon-methane mixtures expanding through a nozzle show a population inversion in the neutral carbon line at 2478.6 Å corresponding to the $2p^2\ ^1S_0 - 2p\ 3s\ ^1P_1{}^o$ transition.

## Theory

In Ref. 1, the possibility of population inversion for a neutral nitrogen atom line at 1745 Å was indicated. Although such a possibility does exist, it is difficult to verify experimentally because the wavelength is within the vacuum ultraviolet range. A line transition similar to 1745NI exists, however, in neutral carbon at 2478.6 Å which is in the quartz-ultraviolet wavelength region. Thus, the present experiment was conducted with this carbon line.

The population inversion for any spectral line can be observed by measuring the apparent absorption coefficient of the line. This is done by placing a concave mirror of reflectance r behind the plasma (see Fig. 1). If $I_1$ is the direct plasma radiance with a mirror $M_1$ covered and $I_2$ is the sum of the direct emission and the partially absorbed emission, then

$$I_2 = I_1 + rI_1 - \Delta I \qquad (1)$$

where $\Delta I$ is the radiance absorbed within the plasma. The line absorption $A_L$ defined as the ratio of the absorbed to the incident radiance[3,4] is given by

$$A_L = \Delta I/(rI_1) = (1 + r - I_2/I_1)/r \qquad (2)$$

Because the spectrograph spectral band pass is usually large compared with the line width, $I_1$ and $I_2$ are radiances integrated over the entire line profile and path length.

The observed line absorption coefficient, Eq. (2), can be related to the population densities of the upper and the lower states of the line concerned as follows. From the radiative transfer theory[3,4] one can show for a homogeneous path of length L that the integrated line absorption $A_L$ is given in terms of atomic parameters by,

$$A_L = \frac{\int_{\text{line}} \left(1 - e^{-\kappa_\nu{}^* L}\right)^2 d\nu}{\int_{\text{line}} \left(1 - e^{-\kappa_\nu{}^* L}\right) d\nu} \qquad (3)$$

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where the integral is performed over the entire line profile. The linear absorption coefficient $\kappa_\nu^*$ is given by

$$\kappa_\nu^* = \frac{\pi e^2}{mc} \frac{N_\ell^*}{\sqrt{\pi}} \frac{f_{\ell u}}{\Delta\nu_D} H(a, u) \tag{4}$$

$$= \kappa_o^* H(a, u)$$

where e = electron charge, m = electron mass, c = velocity of light, $f_{\ell u}$ is the absorption oscillator strength for the transition $\ell \rightarrow u$, $\Delta\nu_D = (\nu_o/c)\sqrt{2kT/M}$ is the Doppler e-folding width for the line centered at frequency $\nu_o$, T is the heavy particle kinetic temperature and M the heavy particle mass. H(a, u) is the Voigt function[5] where $a = (\Gamma/4\pi)/\Delta\nu_D$ is the ratio of the Lorentz damping width to the Doppler width and $u = \Delta\nu/\Delta\nu_D$ is the distance from the line center in units of the Doppler width. $N_\ell^* = N_\ell(1 - (N_u/g_u)/(N_\ell/g_\ell))$ is the lower state density corrected for stimulated emission. The quantity $\kappa_o^*$ is the absorption coefficient at the center of a pure Doppler line. One can transform Eq. (3) to a more convenient form

$$A_L = 2 - \frac{\int_{\text{line}} \left(1 - e^{-2\kappa_\nu^* L}\right) d\nu}{\int_{\text{line}} \left(1 - e^{-\kappa_\nu^* L}\right) d\nu} \tag{5}$$

which can be evaluated in terms of the equivalent width $W_\nu$ defined by

$$\frac{W_\nu}{2\Delta\nu_D} = \int_0^\infty \left(1 - e^{-\kappa_\nu^* L}\right) du$$

For the experimental conditions of interest, $a \ll 1$ so that the Doppler limit for H(a, u) applies, i.e. $H(a, u) = e^{-u^2}$, and $W_\nu/2\Delta\nu_D$ can be expanded in a series[6].

Substituting this series into Eq. (5) we find for $\kappa_o^* L \ll 1$,

$$A_L = \frac{\kappa_o^* L}{\sqrt{2}}$$

For larger values of $\kappa_o^* L$, Mitchell and Zemansky[3] give

$$A_L = \frac{\sum_{n=1}^{\infty} (-1)^{n+1} a_n (\kappa_o^* L)^n}{1 + \sum_{n=1}^{\infty} (-1)^n b_n (\kappa_o^* L)^n} \tag{6}$$

where

$$a_n = [2^{n+1} - 2]/[(n+1)!\sqrt{n+1}]$$

and

$$b_n = 1/[(n+1)!\sqrt{n+1}]$$

Comparing Eq. (6) and (2), we note if $\Delta I > 0$ ordinary absorption processes dominate over the stimulated emission, and $A_L > 0$, whereas if $\Delta I < 0$, a population inversion has occurred and $A_L < 0$.

## Equipment and Measurements

The measurements are conducted in the free-jet stream produced in a constricted-arc plasma-jet wind tunnel of 1.27 cm constrictor diameter at Ames Research Center. The contoured nozzle is approximately 25 cm long and has an exit diameter of 5.95 cm. The measurements are made 11 cm downstream of the exit plane. Since the wind tunnel nozzle operates in a highly under-expanded mode, an abrupt expansion takes place within the 11 cm distance between the nozzle exit plane and the measured station. The area ratio at the test station is approximately 50. The diameter of the plasma-jet stream at the measuring station is of the order of 10 cm, of which approximately 5 cm is considered to contain the hot plasma core. A mixture of helium and methane or argon and methane is used as the working gas, and the methane content is of the order of 5% by mole. The methane is presumed to dissociate within the arc-heater and provide the atomic carbon gas necessary for the described transition. The stagnation chamber (i.e., the cathode chamber) pressure is maintained at around 2 atm and the enthalpy of the mixture varies between approximately 50 and 200 Mj/Kg at the centerline of the jet.

The measurement of $I_1$ and $I_2$ in Eq. (2) is performed by using the mirror-chopper system shown in Fig. 2. As shown in the figure, it consists of two spherical mirrors, a Brower Model 1513 ratiometer system having two phase-locked choppers, two lock-in voltmeters, and the ratiometer, together with a digital voltmeter and a two-channel recorder. This system provides both the separately time-averaged signals $I_1$ and $I_2$ and their ratio. The overall repetition frequency was approximately 20 Hz, with a typical signal averaging time of 3 sec, thus resulting in a well-stabilized signal of high signal-to-noise ratio. The radiance is measured using a 1/2 meter scanning monochrometer having a reciprocal dispersion of 17.5 A/mm. A Hitachi R106 ultraviolet sensitive photomultiplier tube was used as the light detector. Slit widths were varied between 0.05 and 0.3 millimeters.

In addition to the ratio $I_2/I_1$, one must know the mirror reflectance r at 2479 Å in order to determine the absorption $A_L$ by Eq. (2). The value of r is crucial, since a small uncertainty in r will seriously affect the magnitude and sign of $A_L$, as is apparent from the form of Eq. (2). Three different methods are employed to determine the mirror reflectance at the required wavelength.[4] In the first method, which does not require any separate calibration, and can be performed during a run, one observes the optically-thin continuum at wavelengths a few angstroms away from the line. Ionized helium emits a moderately strong free-bound continuum at wavelengths below 2600 Å due to radiative recombination into $2s\,^3S$ state. This free-bound continuum is optically thin under the present experimental conditions and hence $\Delta I = 0$ for the continuum. Equation (2) then gives

$$r = (I_2/I_1 - 1)_{\text{continuum}}$$

The second method of determining r is similar to the first in that it utilizes the emission from the plasma as the light source. Instead of using the helium continuum, however, the intensity ratio $I_2/I_1$, of the 2479 Å carbon line itself was monitored as the carbon mole fraction was increased from $\simeq 10^{-5}$ up to .033. At the lower limit the line is optically thin and the same formula as used to determine r from the continuum intensity ratio is valid.

The third method of determining r involves an independent measurement of mirror reflectance using an ultraviolet light source. A special calibration rig was designed for this purpose having the features shown in Fig. 3. As shown in the figure, it includes an ultraviolet light source (NBS Standard of Spectral Irradiance) and an ultraviolet beamsplitter (transmittance $t_r$ and reflectance $r_r$) each of which can be rotated precisely 90° around the flow axis. Based on the four measurements $I_a$, $I_b$, $I_c$ and $I_d$, shown on Fig. 3, the mirror reflectance r is given by

$$r = I_b \; I_d/(I_a I_c)$$

All three methods yield a value of r = 0.58 at 2479 Å for the mirror $M_1$. The precision of the intensity ratio $I_2/I_1$, set mainly by the arc tunnel unsteadiness, is ± .005.

## Results

Experimental values obtained in both helium-methane and argon-methane plasmas are shown in Table 1. Numbers 1 through 7 are helium-methane runs, while 8 and 9 are argon-methane runs. The largest absolute value of $A_L$ is that for run number 5.

To show that these values of $A_L$ give quite reasonable population inversions, we calculate $N_\ell^*$ and $(N_u/g_u)/(N_\ell/g_\ell)$ for Run 5. Typical pressures and heavy particle temperatures in the test section are 1 torr and 1000°K respectively. Estimates based on the results of Ref. 1 and the appearance of the upper members of the Balmer series (HI (13) is still quite distinct) indicate $T_e = 3500\ ^oK$ and $N_e = 1 \times 10^{14}$ in the test section. The value of $\Delta\nu_D = 4.8 \times 10^9$ hz. The contributions to Γ are: heavy particle collisions $\Gamma_{coll} = 2\nu_{coll} = 1.3 \times 10^6$ hz; Stark broadening $\Gamma_{Stark} = 2\pi\Delta\nu_{1/2} = 3 \times 10^7$ hz ($\Delta\lambda_{Stark} = 10^{-5}$ Å) (see Ref. 7); and radiation $\Gamma_{rad} = A_{u\ell} = 3.4 \times 10^7$ hz[8]. Summing these contributions we find $a = (\Gamma/4\pi)/\Delta\nu_D = 1 \times 10^{-3} \ll 1$. The total particle density $N_{tot}$ is $1 \times 10^{16}\ cm^{-3}$. For the CI 2478.6 Å line, Wiese[8] gives $g_u = 3$, $g_\ell = 1$ and $f_{\ell u} = .094$. From Eq. (6) we find for $A_L = -2.24$ that

$$\kappa_o^* L = -1.46$$

For an assumed path length L = 5 cm, $\kappa_o^* = -.292\ cm^{-1}$. Evaluating Eq. (4) for this transition

$$\kappa_o^* = 2.93 \times 10^{-13} N_\ell^*$$

and hence $N_\ell^* = -1.0 \times 10^{12}\ cm^3$ for Run 5. Note $|N_\ell^*| \ll N_{CI}$. The energy of the lower state for this transition is $E_\ell = 21648\ cm^{-1}$ and hence $N_\ell/g_\ell = (N_{CI}/Z_{CI}) \exp(-E_\ell/kT_e) = 5.01 \times 10^9\ cm^{-3}$ where the neutral carbon partition function $Z_{CI} = 9$ at $T_e = 3500\ ^oK$. The low states are assumed to be in partial equilibrium among themselves[1] at an effective excitation temperature = $T_e$. Using $N_\ell^* = N_\ell(1 - (N_u/g_u)/(N_\ell/g_\ell))$ we find $(N_u/g_u)/(N_\ell/g_\ell) = 2.01 \times 10^2$ for Run 5.

It should be noted that the measured $A_L$ values are averaged across the jet and local values may be more negative.

## Conclusion

Experimental measurements have indicated that a substantial population inversion can be produced in a high pressure arc heated carbon plasma expanding through a nozzle for the carbon I 2478.6 Å line.

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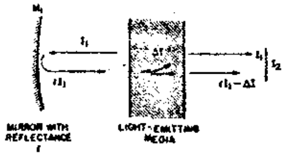

Figure 1. Schematic of single-reflection absorption measurement.

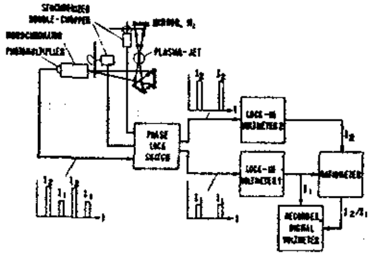

Figure 2. Schematic of $I_2/I_1$ measuring technique.

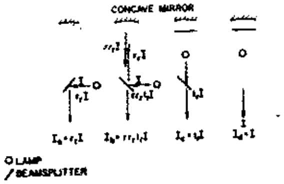

Figure 3. Procedure for determining mirror reflectance. r = reflectivity of concave mirror $M_1$, $r_r$ = reflectivity of beamsplitter, $t_r$ = transmission of beamsplitter, I = radiance of light source.

Table I

| Number | Mole Fraction Carbon | Cathode Pressure atm | Total Mass Flow gm/sec | Enthalpy $M_J/Kg$ | $\frac{\Delta I}{rI_1} = A_L$ | $\kappa_o{}^* L$ Eq. (6) |
|---|---|---|---|---|---|---|
| 1 | .051 | 3.0 | 2.2 | 140 | - .362 | - .42 |
| 2 | .051 | 3.0 | 2.1 | 210 | - .276 | - .33 |
| 3 | .051 | 3.0 | 2:1 | 210 | -1.57 | -1.20 |
| 4 | .067 | 2.72 | 2.1 | 130 | -1.10 | - .96 |
| 5 | .033 | 4.75 | 2.5 | 190 | -2.24 | -1.46 |
| 6 | .080 | 2.38 | 1.9 | 140 | -1.90 | -1.33 |
| 7 | .031 | 1.97 | 1.8 | 140 | -1.10 | - .96 |
| 8 | .03 | 2.05 | 5.0 | 40 | - .845 | - .80 |
| 9 | .03 | 3.05 | 6.0 | 50 | - .845 | - .80 |

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